A Study on Characterization of Aerosol and Estimation of Direct Radiative Forcing over Important Cities in Indo-Gangetic Plain

A Synopsis

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INTRODUCTION

Over last decade, there has been increasing interest of scientists and researchers to explore effects of aerosols on climate and human health. Aerosols are light particles suspended in the air and they easily carried over by the wind from one place to another which results in regional environmental effects. Aerosols have potential to change chemistry in the troposphere and stratosphere (Ravishankara, 1997; Satheesh et al., 1999) which significantly influence global environment. High spatial and temporal variability of aerosols make assessment and prediction of its effects on environment and health more challenging (Moorthy, Satheesh, & Murthy, 1997; Satheesh & Krishna Moorthy, 2005). Large part of total aerosols comes from the natural sources and small but significant produced anthropologically near or in human habitats which effect humans in a great deal (Ramachandran, Srivastava, Sumita, & Rajesh, 2012). Aerosol size spans five orders of magnitude which varies from 10^{-3} to 10^{2} μm, a size more or less depends on the source. On the basis of size, aerosols are generally classified into three following categories (1) Nucleation mode or Aitken mode (radius ~ 0.001 to 0.1 μm) (2) Accumulation mode (radius ~ 0.1 to 1.0 μm) and (3) Coarse mode (radius >1 μm). The nucleation mode aerosols are produced mainly by the nucleation of volatile gases, a process known as, gas–to–particle conversion, in the atmosphere; the accumulation mode by coagulation (a process in which two or more aerosols combine together to form a single larger aerosol) and growth of nucleation mode aerosols by condensation of water vapors and coarse mode aerosols directly by mechanical processes (e.g., action of winds on aerosols) (Steinfeld, 1998). Aerosols occur with different shapes (Reist, 1984). Nevertheless, broadly they can be classified as follows: (i) Isometric particles are those for which all three dimensions are roughly the same like spherical particles. (ii) Platelets are particles that have two long dimensions and one small third dimension like leaf fragments. (iii) Fibers are particles with great length in one dimension and much smaller dimensions in the other two like threads or mineral fibers.

The sources of atmospheric aerosols can be both natural and anthropogenic. The aerosols of terrestrial origin are formed generally by two mechanisms (i) mechanical disintegration processes (primary sources) and (ii) gas-to-particle conversion processes (secondary sources). On a global scale natural sources are stronger in abundance compared to anthropogenic sources, but on a regional scale anthropogenic sources can be stronger especially near industrialized regions (Ginoux, Prospero, Gill, Hsu, & Zhao, 2012). Natural sources include production from sea spray, dust produced from soil by winds and volcanic eruptions. Anthropogenic sources include industrial emissions, transportation and bio mass burning (Kumar et al., 2010). The composition of aerosols and particles depends on their source. Wind-blown mineral dust tends to be made of mineral oxides and other material blown from the Earth's crust; this particulate is light-absorbing. Aerosols produced from different natural and anthropogenic activities are mixed together and hence each aerosol particle is a composite of different chemical constituents (Satheesh & Krishna Moorthy, 2005). Chemical composition of aerosols determine their complex (contain real and imaginary parts) refractive index. Particle refractive index is an important parameter while determining its radiative effects. The real part determines its scattering properties and imaginary part, the absorption characteristics (Steinfeld, 1998). The chemical composition and hence refractive index depends on the source of particles. The real part of particle refractive index usually lies in the range 1.3 to 1.6 and imaginary part varies over several orders of magnitude from about 5×10^{-9} to 5×10^{-1}. Particles originating from combustion (burning) processes usually have high absorption properties and hence high imaginary part of refractive index. Vehicular emissions, fossil fuel burning, wood fire and industrial emissions impact local environment (Fenger, 1999; Mayer, 1999) and have serious health issues (Harrison & Yin, 2000; Pope et al., 2002).

Aerosols particles play an important role in radiation budget of the atmosphere. Because aerosols are regionally concentrated, absorbing aerosols can impose a large horizontal gradient

in radiative heating, which can directly alter the general circulation, which in turn can perturb the precipitation patterns (Russell, Hobbs, & Stowe, 1999). Aerosols modify incoming solar and outgoing IR radiation. Change in radiation flux caused by aerosol is referred to as aerosol radiative forcing (Pandithurai et al., 2008). The effect of aerosols on the top of the atmosphere (TOA) radiative fluxes is called top of the atmosphere radiative forcing and that on surface radiative fluxes is called the surface radiative forcing (Satheesh, Vinoj, & Krishna Moorthy, 2010). The difference between the two is the atmospheric radiative forcing. Aerosol forcing is defined as the difference in radiative fluxes with and without aerosols.

$$\text{Aerosol Forcing } (\Delta F) = \text{Flux}_{\text{without_aerosol}} - \text{Flux}_{\text{with_aerosol}}$$

Radiative forcing is usually expressed in units of W/m^2, and can be a positive or negative term. A reduction of solar radiation reaching the earth is considered a negative forcing. Aerosols both absorb and scatter solar radiation. Most of the light extinction caused by aerosols is due to scattering. Aerosols not only influence the incoming solar and outgoing infrared radiation but also influence the size, abundance, and rate of production of cloud droplets. They also act as cloud condensation nuclei (CCN) (Sun & Ariya, 2006). Thus they influence cloud cover, cloud albedo, and cloud lifetime.

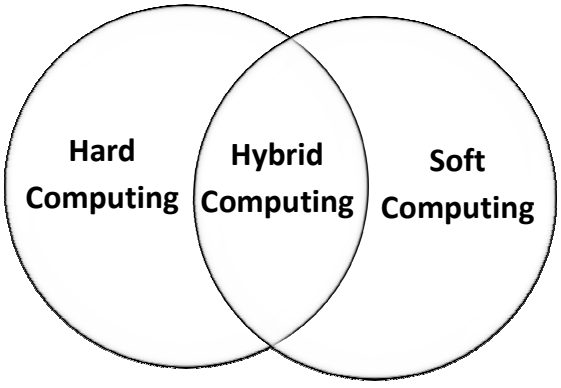
Effects of aerosols on solar radiation can be broadly classified into two Categories; (1) Direct Radiative forcing and (2) Indirect Radiative forcing.

Direct Radiative Forcing: The interactions of radiation with aerosols can be either on account of scattering or absorption. The scattering and absorption of visible radiation (short wave) by aerosols produces changes in climate by changing the planetary albedo. Absorption and re-radiation of IR enhances the atmospheric greenhouse warming. These two effects are called direct forcing of aerosols on radiation and climate. The presence of aerosol increases the surface reaching IR radiation and decreases the outgoing IR radiation. Since most of the aerosols are

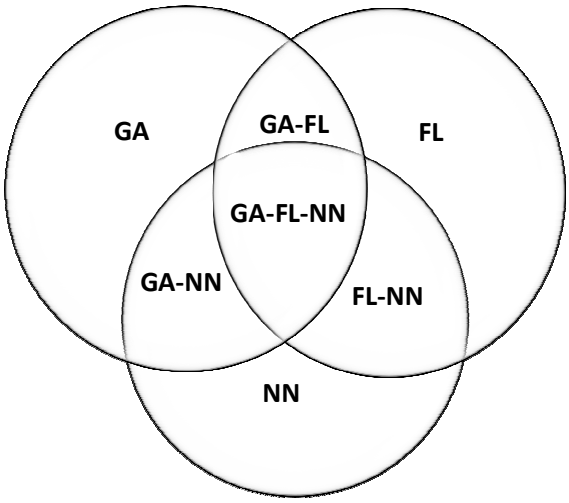
concentrated near the surface, absorbing aerosol component heat the lower atmosphere (Shekar Reddy & Venkataraman, 2000). The direct radiative effect of aerosols is also very sensitive to the single scattering albedo ω. If change in ω from 0.9 to 0.8 can often change the sign of the direct effect, depending on the albedo of the underlying surface and the altitude of the aerosols (Hansen, Sato, & Ruedy, 1997). Recent estimates for the direct forcing by anthropogenic aerosols are between -0.3 to -0.9 Wm^{-2} for sulphate aerosols, between -0.15 and -0.25 Wm^{-2} for biomass aerosols (from biomass fuel use and land clearing), and between +0.15 and +0.2 Wm^{-2} for fossil fuel and industrial sources of organic and black carbon aerosols (Penner, Chuang, & Grant, 1998). Thus, the climate forcing associated with anthropogenic aerosols could offset a substantial fraction of the warming associated with greenhouse gases (Tegen, Lacis, & Fung, 1996).

Indirect Radiative Forcing: Aerosols can act as cloud condensation nuclei (CCN) for the formation of clouds. An increase in the concentration of aerosols can result in an increased concentration of cloud droplets, which in turn can increase the albedo of clouds (Reist, 1984). This causes a decrease in the visible solar radiation reaching the surface. Thus cloud albedo has a significant role in determining the global energy balance. An increase in aerosols increases the cloud droplet concentration and reduces the mean droplet size. This is because the water vapour availability per aerosol is less when the aerosol number is more. This increases the cloud lifetime and thus inhibits precipitation. Changes in droplet concentrations due to carbonaceous aerosols from biomass burning and fossil fuel/industry lead to estimates of forcing that range from –1.2 to –1.6 Wm^{-2} depending on the magnitude of natural organics assumed in the calculation (Penner et al., 1998). Estimation of aerosols effect on radiation is more uncertain than that due to well mixed green house gases, because of their short life times, highly inhomogeneous spatial distribution and its complex nature of interaction with radiation (Hansen et al., 1997).

Soft computing is a set of methodologies that seek to develop the patience for imprecision and uncertainty to achieve tractability, robustness, and low solution cost. Soft computing techniques such as Fuzzy Logic (FL), Neural Networks (NN), Genetic Algorithm (GA), Ant Colony Optimization (ACO), and Particle Swarm Optimization (PSO) have received a lot of interest of researchers due to their potentials to deal with highly nonlinear, multidimensional, and ill-behaved complex engineering problems (Tettamanzi, Janßen, & Tomassini, 2013). In fact, soft computing's main characteristic is its essential capability to create hybrid systems that are based on the integration of constituent technologies (Tettamanzi et al., 2013). Hybrid computing is the combination of two major problem-solving technologies includes: hard computing and soft computing. Hard computing deal with precise models where accurate solutions are achieved quickly. On the other hand, soft computing deals with approximate models and gives solution to complex problems.



1a



1b

Figures: 1. 1a) Hybrid computing scheme and 1b) Shows schematic diagram of intersections of members of soft computing family

Soft computing techniques have become one of promising tools that can provide practice and reasonable solution. Soft computing (SC) techniques are used in different fields and some of application areas of soft computing are shown in figure 2.

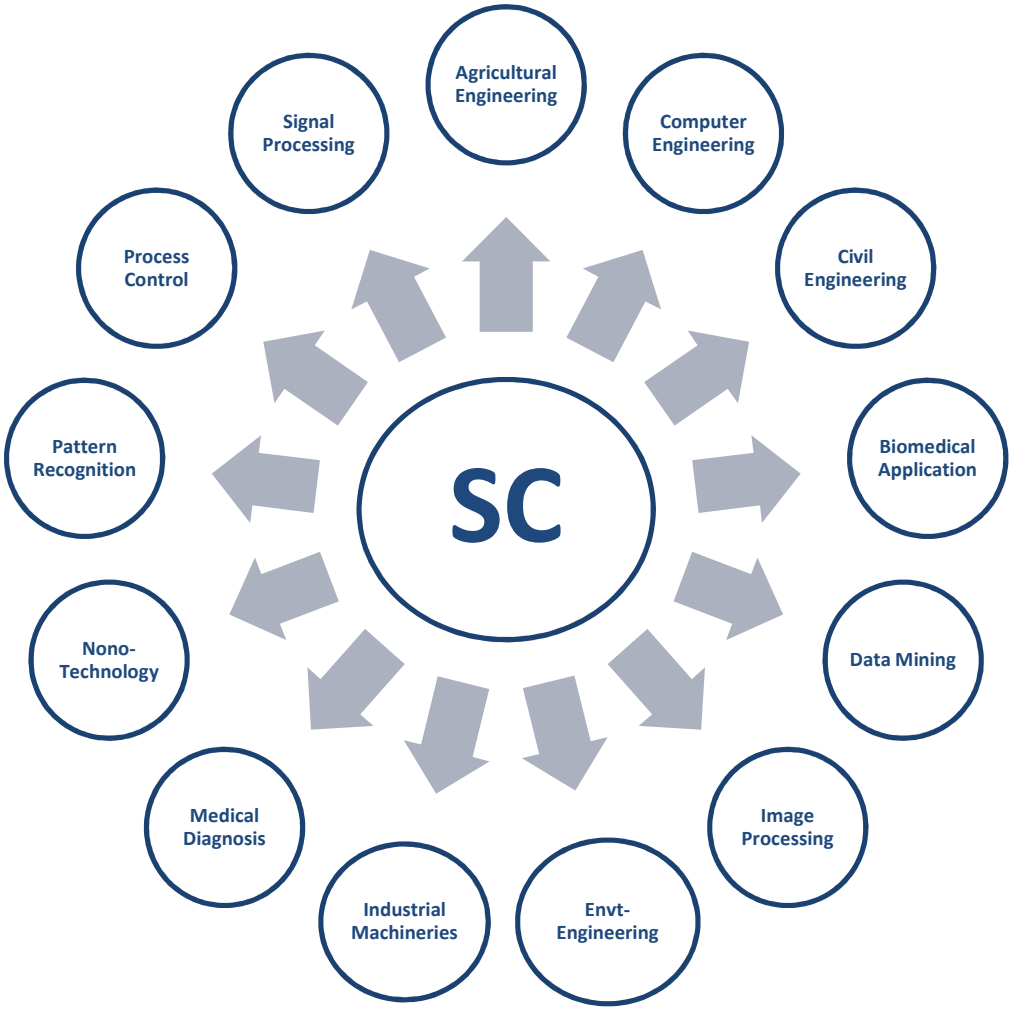


Figure: 2 Application areas of soft computing.

Optical properties of aerosols depend on the aerosol particle chemical constituents. Refractive index of aerosol particle is in general complex number which depends on its chemical constituents. The real part determines its scattering properties and imaginary part, the absorption characteristics. Optical properties of atmospheric aerosols such as the aerosol optical depth (AOD), scattering coefficient, single scattering albedo (SSA), extension coefficient and the

phase functions as well as information on their spectral dependencies (Tiwari et al., 2015), are quite important to compute the radiative impacts and regional climate forcing of aerosols (Li et al., 2014). So accurate estimation optical properties are very important to compute radiative forcing (Ram, Singh, Sarin, Srivastava, & Tripathi, 2016).

The study of aerosol is important for many reasons. It is thought that aerosol may be involved in a feedback to global warming. It is certainly important in the Earth's radiation budget. There are also concerns about the effects of aerosol on human health. Aerosols, in some cases, an important part of the chemical deposition budget for chemical species to ecosystems. Knowledge of aerosol properties is essential for correcting the atmospheric effect in satellite remote sensing of Earth's surface.

MOTIVATION

The Ganga basin or the Indo-gangetic basin (IGB) plain hosts more that 40% of the total population of India. IGB houses more than 600 million people and is the most populated river basin in the world (Sen et al., 2016). Ganga basin spans about 2000 km in length along North-West to South-East and has more and less 400 km wide which is bounded by the Himalaya. Through satellite data and ground base studies by many researchers it is very well established fact that the Ganga basin comes under one of the highest aerosol load sites throughout the year. In terms of aerosol loading, IGB is one of the most polluted regions in the world (Jethva, Satheesh, & Srinivasan, 2005). In Ganga basin a major part of Indian population experiencing the critical effect of aerosol on the environment and facing many health issues caused by aerosols. Important cities in indo-Gangetic Basin have been selected as the sites for the present study and five three of UP, namely, Agra, Allahabad, Kanpur, Varanasi, other Delhi NCR, Kolkata and Hisar region have been chosen as the sites for the data comparison with Agra site.

OBJECTIVE

The present research objective is to address direct effects of aerosol on the climate and the radiative forcing due to the aerosol loading over important cities in Indo-gangetic basin (IGB). Agra, the site of interest has cultural and heritage importance as well as comes under favorable conditions to study accumulated and transported aerosols because site is bounded in North by the Himalayas and in the west Thar Desert. The main emphasis of proposed research objective is to study aerosol loading over the site of interest, identification of sources, assessment of aerosol loading over radiative forcing and effect of black carbon on radiative forcing. To handle proposed problem in efficient manner the problem is specified in terms of specific objectives given below:

Specific objectives are:

1. *To measure aerosol (PM_{10}, & $PM_{2.5}$) mass concentration over Agra region.*
2. *To measure aerosol optical depth and study its seasonal variation.*
3. *To compute aerosols optical properties using measured physical parameter of aerosol.*
4. *To find the correlation between measured AOD and satellite data.*
5. *To compute direct radiative forcing at the bottom of atmosphere and top of atmosphere over important cities in Indo-Gangetic Plain.*
6. *To explore possibilities of fast and accurate computation of optical properties and radiative forcing using soft computing.*
7. *To develop graphical user interface for efficient computation of radiative forcing.*

METHODOLOGY

- **Site Description**

For ground base observations, Agra has been selected as investigation site. Agra, a site in the Ganga basin, which is famous for the Taj-Mahal and cultural heritage has a high load of suspended particulate matter SPM. A number of studies have been carried out in Agra in the last one decade, but all were on aerosol chemical characterization. Modeling and measurements of optical properties of aerosol and an assessment of its effects on radiative forcing have not been attempted by any researchers. The city has many small-scale and cottage industries in its periphery. Agra, located on the Indo-Gangetic Plain has a continental sub-tropical climate, with long, hot summers from April to September when temperatures can reach as high as 45°C (113°F). During summers dry winds blow in this region. The monsoon months from July to September see about 67cm (27 inches) of rainfall annually. The climate in Agra offers great variations season to season, with extremely hot weather, periods of heavy rainy weather, and cooler spells during the winter months. Agra's winter climate roughly falls between October and January, where rainfall levels are low and average daytime temperatures in winter stay around 22°C or more. The weather in Agra is at its sunniest and hottest between April and July, when the climate quickly heats up. Tropical temperatures regularly exceed 35°C and even top 40°C at times. Although the level of rainfall increases sharply during this period to over 600 mm / 24 inches, much of the day is dry and the rainy weather usually arrives for just a few hours in the afternoon. Evenings are often fine and the climate feels especially pleasant after a spell of heavy rain.

The IGB compared to other regions in India experiences four distinct seasons, winter (DJF) combining Dec-Feb, summer (MAM) combining Mar-May, monsoon (JJA) combining Jun-Aug and post monsoon (SON) combining Sep-Nov.

Dayalbagh: Dayalbagh area is located towards the north of Agra city. The place constitutes an extension of the city. The place lies at a distance of about 3 km form NH-2. Although it forms the peripheral regions of the city but the place has a good influence of the city.

Original name: Dayalbagh

Geographical location: Agra, Uttar Pradesh, India, Asia

Geographical coordinates: 27° 13' 0" North, 78° 1' 0" East

Site Description of other cities of interest:

Allahabad city is among the largest cities of Uttar Pradesh and situated at the confluence of three rivers - Ganga, Yamuna and the invisible Saraswati. The meeting point is known as Triveni and is especially sacred to Hindus. The geographical coordinates of Allahabad are 25°26′56″N, 81°49′59″E, 104 m elevation (above mean sea level). The city of Allahabad is located in south east part of Uttar Pradesh State in the IGP, and has population of over >1 million, population density of ~17,000 km^{-2} and is located very close to Kanpur. Allahabad experiences wide range of temperature which goes as high as ~46^0C in summer to a low as ~1^0C in winter. The major source of emission in the locality is the automobiles. Other sources include scattered small and medium industries and tanneries. The usual practice of open fireplaces by local people during winter also adds to the air pollution on a large scale.

Kanpur is the largest city in the state of Uttar Pradesh in India. It is the administrative headquarters of Kanpur Nagar district and Kanpur division. It is the second largest industrial town in north India, following Delhi. Kanpur is known as the Leather city of the world. It is also called Manchester of the East. The geographical coordinates of Kanpur are 26°26′52″N, 80°20′46″E, 142 m elevation (above mean sea level) an urban/industrial site, is located in the central part of the IGB and earlier studies have confirmed the huge aerosol load over this region. The high aerosol loading not only influences the cloud microphysical properties over Kanpur, but over the entire IGB. Previous studies have assessed the seasonal variability of the

aerosols in this region, the influence of the dust events on the aerosol optical properties and estimated the aerosol DRE (Direct Radiative Effect) for only few months duration. However, none of them addressed how seasonal variations in aerosol optical properties affect the regional climate, how clouds might affect the aerosol DRE in this region, and how much the contribution of anthropogenic fraction to the composite aerosol DRE could be.

Varanasi city is an ancient center of Indian culture and religion. It is a revered city for all Hindus and Buddhists worldwide. The city has relatively high population density. The geographical coordinates of Varanasi are $25^{\circ}20'59.23''N$, $82^{\circ}58'44.67''E$, 82.93 m elevation (above mean sea level). The site receives the air pollution from the vehicular activities and from nearby cottage industrial sources. Here also the traffic volume is reasonably high with mostly local transport vehicles.

Delhi city is the capital of India. The experimental site Delhi is typically represents the plains of Ganga basin in the northern India. The geographical coordinates of Delhi are $28^{\circ}39'13.00''N$,$77^{\circ}13'44.00''E$, 229 m elevation (above mean sea level). Delhi is among the ten most polluted cities in the world and the second largest Indian megacity with an average population growth rate of 3.85% per year (A. Srivastava & Jain, 2008). Consequently, vehicular growth rate on an average is 5.85% per year (Economic Survey of Delhi 2006). This alarming vehicular growth rate has resulted in a significant rise in the TSPM (total suspended particulate matters) level over Delhi. Although the vehicles are the biggest contributor to the ambient TSPM level, significant contributions from other sources such as industries, roadside dust, trans-boundary migration, power plants and local sources have also been observed (A. Srivastava & Jain, 2007). The experimental site is located in the heart of Delhi and no major industries are located within 5 km radius around.

Kolkata city is the capital of the Indian state of West Bengal which is located on the east bank of the Hooghly River. The geographical coordinates of Kolkata are $22^{\circ}33'45.00''N$,$88^{\circ}21'46.00''E$,11 m elevation (above mean sea level). Kolkata has a Tropical wet-and-dry climate climate, with summer monsoons.

Hisar district is one of the 21 districts of Haryana state, India. Hisar pronunciation previously spelled Hissar, is the administrative headquarters of Hisar district in the state of Haryana in northwestern India. It is located to the west of New Delhi, India's capital, and has been identified as a counter-magnet city for the National Capital Region to develop as an alternative center of growth to Delhi. The geographical coordinates of Delhi are $29^{\circ}08'56''N$,$75^{\circ}44'12''E$, 212 m elevation (above mean sea level).

- **Mass Concentration**

Mass concentration or load of aerosols:

The total mass of particles per unit volume of air is one of the major parameters used to characterize particles in air and along with size, is the basis of air quality standards for particulate matter.

RSPM :

Respirable Suspended Particulate Matter (RSPM) is particles of size upto 10 µm which are inhalable. RSPM can be collected on Whatmann GFA filter paper using single stage PM_{10} aerosol sampler (APM 450 Envirotech High Volume PM_{10} Sampler) (Goyal, Chan, & Jaiswal, 2006). PM_{10} aerosol samples can be collected at a flow rate of 0.7-1.1 m^3 min^{-1}. Particles deposited on filter after sampling for a certain period at a certain flow rate can be determined gravimetrically. The difference in the mass of filter paper after the sampling and before the sampling is PM_{10} load (Querol et al., 2001).

$PM_{2.5}$:

The aerosol particles in the smaller size range ≤ 2.5 µm ($PM_{2.5}$) are of great interest with respect to health effects (Sharma et al., 2014). $PM_{2.5}$ samples can be collected on PTFE filters using Envirotech/Polltech $PM_{2.5}$ samplers (Zheng et al., 2005). The difference in the mass of filter before the sampling and after the sampling gives $PM_{2.5}$ loads (Querol et al., 2001). From the mass of the particulates collected and the volume of the air sampled, the mass concentration of the suspended particulates in the ambient air can be computed as µg m^{-3} of air. Mass of aerosol (in µg m^{-3}) = mass of the aerosol deposited on filter paper/total air volume Where, mass of aerosol deposited on filter paper is difference in the wt. of filter paper before the sampling and after the sampling and total air volume (m^3) is multiplication of flow rate (m^3 min^{-1}) and sampling periods (minutes).

Number concentration of aerosol and Number size distribution of aerosol:

The number concentration of aerosol and number size distribution of aerosol is important parameters to characterize the aerosol (Stanier, Khlystov, & Pandis, 2004). Condensation nucleus counters (also referred to as condensation particle counters or Aitken Nuclei Counters: CNCs, CPCs, ANCs) measure the total aerosol number concentration larger than some minimum detectable size. . CNCs can detect individual particles as small as 0.003 µm (10~20 g), so they provide an extraordinarily sensitive means for detecting small amounts of material.

The particle size analyzer/dust monitor (Grimm, Germany) or optical particle counter (OPC) can be used for the continuous measurements of number of particles in the air. The instrument measures the cumulative number concentration in 15 channels in the range of 0.3 to 20 µm with a detection range in 1-10^6 counts $liter^{-1}$ with a sensitivity of 1 particle $liter^{-1}$. Particle number distribution can also be measured.

Counting the particles and measuring the sizes can be done by optical microscopy (for particles with diameters from about 0.4 µm to several hundred microns) or electron microscopy (for particles from about 0.001 µm and larger).

AOD/Water Vapour measurement at agra

Aerosol Optical Depth (AOD)

In assessing the radiative forcing due to aerosols over a given region, the single parameter of highest importance is the spectral aerosol optical depth (AOD), which is the integration of the extinction coefficient over a path length through the atmosphere (Babu, Gogoi, Kumar, Nair, & Moorthy, 2012). The optical thickness or depth (τ), for a path length x is defined by

$$AOD = \tau = \int_0^x \beta(x)\, dx$$

Which implies that the coefficient β, (a constant which defines the ability of a unit volume of a suspension to scatter light of a specified wavelength totally), has a constant value over the distance *x*.

During Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB), regular measurements of AOD were made using three different instruments; the 10-channel multi-wavelength radiometer (MWR) and the 5-channel microtops sun photometer. The MWR (Moorthy & Satheesh, 2000; Moorthy et al., 1997) estimated AODs at ten wavelength bands centered at 380, 400, 450, 500, 600, 650, 750, 850, 935, 1025 nm. The Microtops (Solar Light Company, USA) is a hand-held sun photometer that provides AOD and columnar water vapour from instantaneous measurements, based on its internal calibrations, at five channels (340, 500, 670, 936 and 1020 nm) (A. K. Srivastava, Tripathi, Dey, Kanawade, & Tiwari, 2012).

- **Satellite data**

Satellites sensors receive the solar light reflected from the atmosphere and Earth's surface (ocean and land). The light received by the sensor is the sum of light reflected from the surface, light scattered by air molecules and light scattered by aerosols. Over the ocean the reflectivity is

more or less uniform. By knowing the information on the back scattering of air molecules, the aerosol information can be inferred. For mapping aerosol properties on a regional or global scale, satellite based remote sensing is the only option. The most basic aerosol characteristics retrieved from any single wavelength measurement is the extinction or aerosol optical depth. Many satellites data are available for aerosol optical depth. In the proposed study, we intend to use MODI satellite data (King et al., 2003) for comparison of ground based and satellite scene observations.

Computation of optical properties using OPAC for IGP

Optical properties of aerosols depend on the aerosol particle chemical constituents. Refractive index of aerosol particle is in general complex number which depends on its chemical constituents. The real part determines its scattering properties and imaginary part, the absorption characteristics. Accurate estimation of optical properties is very important to compute radiative forcing.

Optical Properties of Aerosols and Clouds (OPAC)

Optical Properties of Aerosols and Clouds (OPAC) is a software package with large data of almost all type of aerosol and cloud optical properties in the solar and terrestrial spectral range. The data are provided in ASCII files and the enclosed software is as FORTRAN program. The FORTRAN program allows reading these files, to mix aerosol components to aerosol types consisting of several components and to calculate resulting optical properties which are not stored in the ASCII files. The optical properties of aerosol are calculated using Optical Properties of Aerosols and Clouds (OPAC) model by incorporating the experimentally observed value of chemical constituents of particulate matter, soot particle (black carbon) and relative humidity of the concerned period. As per the Hess et al., (Hess, Koepke, & Schult, 1998).

- **Calculation of radiative forcing over Agra using SBDART**

Radiative Transfer Model (SBDART)

Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) (Ricchiazzi, Yang, Gautier, & Sowle, 1998) is a FORTRAN computer code designed for the analysis of a wide variety of radiative transfer problems encountered in satellite remote sensing and atmospheric energy budgets studies. The program is based on a collection of highly developed and reliable physical models, which have been developed by the atmospheric science community over the past few decades. SBDART model atmospheres have been widely used in the atmospheric research community and provide standard vertical profiles of pressure, temperature, water vapor and ozone density. In addition, own model atmosphere can be specified. SBDART can compute the radiative effects of several common boundary layer and upper atmosphere aerosol types. The radiative transfer equation is numerically integrated with DISORT (Discrete Ordinate Radiative Transfer). SBDART is configured to allow up to 40 atmospheric layers and 40 radiation streams (40 zenith angles and 40 azimuthal modes). Santa Barbara distort aerosol radiative (SBDART) model is used for calculating aerosol radiative forcing(Ricchiazzi et al., 1998).

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